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(78)

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1. Your reference

P.76032A

2. Patent application number  
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11 2 APR 1999

**9908313.1**

3. Full name, address and postcode of the or of each applicant (underline all surnames)

Rhone-Poulenc Agriculture Limited,  
Fyfield Road,  
Ongar,  
Essex. CM5 0HW.

5683206001

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

United Kingdom

7638987001

4. Title of the invention

WEED CONTROL

5. Name of your agent (if you have one)

J A KEMP & CO

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

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GRAY'S INN  
LONDON WC1R 5LX

26001

Patents ADP number (if you know it)

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Country

Priority application number  
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Date of filing  
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Number of earlier application

Date of filing  
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Yes

- a) any applicant named in part 3 is not an inventor, or
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Patents Form 1/77

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Description 22

Claim(s) 2

Abstract 1

Drawing(s)

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Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents (please specify)

11.

I/We request the grant of a patent on the basis of this application

Signature

Date 12 April 1999

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S. BENTHAM  
0171 405 3292

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## WEED CONTROL

This invention relates to a method for controlling the growth of weeds by the progressive application to, or release into, the surface layer of a growing medium, of  
5 a benzoylisoxazole herbicide, and to compositions for use in the method.

### Background of the invention.

The use of isoxazoles for controlling weeds has been described in European  
10 Patent Publication Nos. 0418175, 0487357, 0527036 and 0560482. The herbicidal activity of diketonitriles (DKN) which may be formed from the isoxazoles has also been described in European Patent Publication Nos. 0213892, 0496630 and 0496631, and International Publication No. WO 95/25099.

Following the application of isoxazole herbicides they may decompose to  
15 diones, in particular diketonitrile (DKN) compounds. This conversion is generally irreversible. The DKN compounds are generally also herbicides. They are generally more water soluble than the isoxazole herbicides and may be subject to movement in the soil profile following rainfall.

It has been found that by modifying the way isoxazole herbicides are applied  
20 the ratio of isoxazole to DKN in a growing medium such as soil can be altered in favour of the isoxazole, increasing the ratio of isoxazole to DKN.

It has been found that by maintaining the ratio of isoxazole to DKN, in favour  
of the isoxazole, in the surface layer of the soil, for example, during the period from application of isoxazole to establishment of a crop, improved control of weeds may  
25 be obtained. Furthermore, crop selectivity may be improved and the risk of run off and leaching may be reduced.

An object of the present invention is to provide a method of application  
and/or a composition which reduces the net movement of isoxazole and DKN  
through the soil and retains the compounds in the soil surrounding, preferably  
30 immediately surrounding, the point of application of the isoxazole.

Another object of the present invention is to provide a method and/or  
composition which permits the delivery of lower individual dose rates of isoxazole

herbicides while maintaining (and sometimes improving) herbicidal efficacy.

A further object of the present invention is to provide a composition, comprising an isoxazole, with improved activity on weed species and/or improved crop selectivity.

5           The objects of the invention can be achieved on whole or in part by the present invention.

It is known that isoxazoles exert their herbicidal activity in plants by conversion to DKN compounds. It might be expected, therefore, that application of isoazole in such a way as to accelerate or favour its conversion to DKN would be  
10           advantageous. The Applicants have found that the opposite is true.

The present invention provides a method for controlling the growth of weeds at a locus in a solid growing medium which comprises treating the locus with a composition comprising an isoxazole herbicide to provide progressive or sequential delivery or release of isoxazole herbicide into the surface layer of the medium.

15           The growing medium includes composts but is preferably the soil.

The locus is preferably a crop-growing locus, for example, where a crop is sown and cultivated.

The surface layer is generally from the surface to a depth of 10 cm, preferably to a depth of 5 cm, more preferably to a depth of 3 cm..

20           According to a feature of the invention the method comprises applying to the locus, for example where a crop is sown and cultivated, sequential low doses of isoxazole herbicides. For example the normal dosage may be divided into two or more, for example 2 to 5, generally equal portions and applied at time-spaced intervals, each application after the first being made, for example 1 to 4 days,  
25           preferably 1 day, after the preceding one.

According to a further feature of the present invention the method comprises treating the locus with a delayed release composition comprising the isoxazole herbicide.

30           The delayed release composition, which constitutes a feature of the invention, may comprise, for example, an encapsulated composition comprising the isoxazole itself or a composition containing it. The delayed release compositions may be prepared by known methods.



The encapsulated product may have a solid outer wall, said wall comprising an inert material, generally having no substantial herbicidal activity.

The encapsulated isoxazole according to the present invention may comprise granules comprising an isoxazole derivative of formula (I), each of these granules being encapsulated with a solid film comprising an inert material itself having no substantial herbicidal activity.

Preferably the inert material is a water-soluble polymeric material, modified by treatment to render it substantially water insoluble.

Soluble materials which may be used include:

a copolyester; polyvinylalcohol; polyacrylate; polycarboxylate; gelatine; polysulfonate, for example the polystyryl polysulfones, a protein, a polyethylene oxide; a modified or unmodified starch; a cellulose for example carboxymethyl cellulose; a dextran, maltose, an alkyl-, hydroxyalkyl-, carboxyalkyl-cellulose; a polyvinylether; poly(2,4-diethyl-6-triazolylethylene); poly(vinylsulfonic acid), polyanhydride, a low molecular weight urea-formaldehyde resin, a low molecular weight melamine-formaldehyde resin, a polymethacrylate for example poly(alkylcyanoacrylate), poly(isobutylcyanoacrylate), poly(2-hydroxyethylmethacrylate), polyacrylic acid or a homologue thereof; low molar mass amphiphiles; low molar mass polymeric amphiphiles; polylactic acid glutamic acid; dendrimers (hyperbranched polymers); phospholipids for example distearoylphosphatidyl choline, dioleoylphosphatidylethanolamine, dipalmitoylphosphatidylchlorine, dipalmitoylphosphatidylglycerol, phosphatidylethanolamine, phosphatidylinositol; lipoprotein, semi-solid poly(ortho ester) polycarboxylates; hydrogels. The materials may be in the form of, for example, solid lipid nano/micro spheres; polyester microspheres; nanocapsules, niosomes; liposomes, polymeric micelles. An oil may be used to facilitate the production of an emulsion with small particle sizes and to inhibit agglomeration.

Preferably the water-soluble material is a copolyester, for example gerol which is a copolymer for example : 1,3-Benzenedicarboxylic acid, 5-sulfomonosodium salt, polymer with 1,3-benzenedicarboxylic acid, 1,4-benzenedicarboxylic acid, 1,2-ethanediol, 2,2'-[1,2-ethanediylbis(oxy)]bis[ethanol] and 2,2'-oxybis[ethanol]. The water soluble materials may vary in molecular weight

and may include oligomers.

The inert water soluble polymer is generally precipitated by association (by complexation or mixing) with a material which does not itself solubilise the aforementioned water soluble polymer. The materials which result in precipitation of the polymer include soluble salts of alkaline earth metals (for example calcium). The association can be modulated by adjusting the pH of the water soluble solution of the polymer which solubilises the ions (of the aforementioned alkaline earth) which effect precipitation of the now insolubilised polymer to encapsulate the particles of the active material. The pH can be adjusted using, for example, acetic acid.

Precipitation can also be induced by adjusting the solvent or solvents without the need for association with another material.

The size of the granules of the active material of an isoxazole derivative of formula (I) is generally from 0.1 to 50 $\mu$ m, preferably from 1 to 20 $\mu$ m.

The thickness of the coating of the encapsulating material is generally from 0.1 to 50 $\mu$ m, preferably from 1 to 20 $\mu$ m.

The granules of encapsulated 4-benzoylisoxazole derivative of formula (I) according to the present invention may be for example, in a powdered state or in a liquid or solid formulation, contained within a support (or a carrier for application).

Compositions of the present invention improve the release of a herbicide to the soil site to which it is applied and movement resulting from rain water or irrigation is reduced. A marked improvement in the downward transmigration of the active ingredient from the immediate application area (weed seed zone) through the soil profile is provided.

Compositions of the present invention provide a method for controlling the release of a herbicide in a range of soil types and edaphic conditions by modification of the ratios of isoxazole: carrier material.

The use of compositions of the present invention permits more efficient use of herbicide which is retained in the area of soil application, for example the weed seed zone; thus the amount of herbicide applied may be reduced. In addition, the herbicide is maintained in the area of the weed seed zone for a longer period of time than usual.

Localisation of the herbicide in the weed seed zone improves selectivity on

the crop species, for example maize.

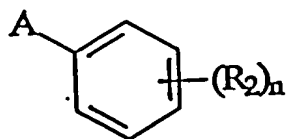
Compositions of the present invention provide a method for reducing the release rate of herbicides, such as isoxazoles, in soil thereby slowing the overall rate of degradation. Encapsulation in a matrix carrier increases the stability of the herbicides as it is protected from the components which may promote degradation, such as moisture or microbial activity.

By the term "pre-emergence application" is meant an application to the soil in which the weed seeds or seedlings are present before emergence of the crop. One example of a pre-emergence application is known as "pre-plant incorporated" (PPI), where the herbicide is incorporated into the soil before planting the crop. Another is where the herbicide is applied to the soil surface after sowing the crop. By the term "foliar activity" is meant herbicidal activity produced by application to the aerial or exposed portions of the weeds which have emerged above the surface of the soil.

In general, the application rate of 4-benzoylisoxazole herbicides of formula (I) in compositions of the present invention is from 0.005kg to 0.5kg herbicidally active compound, preferably from 0.015kg to 2kg herbicidally active compound, more preferably from 0.02kg to 0.12kg herbicidally active compound, even more preferably from 0.05 to 0.09kg herbicidally active compound per hectare. When sequential low doses of isoxazole herbicide are used, as hereinbefore described, the application rates given above may be divided.

The method of the invention is generally applied to a locus pre-emergence of the weeds and crop plant. Preferably the locus is first cultivated and/or treated to remove existing weeds. For example a burn down herbicide such as glyphosate may be used.

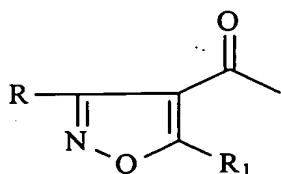
Representative herbicides whose mobility in soil is controlled by compositions of the present invention include 4-benzoylisoxazole derivatives of general formula (I):



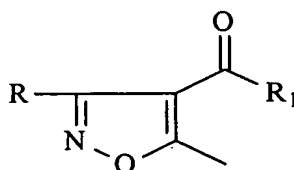
(I)

wherein

A represents a group (A-1) or (A-2):



(A-1)



(A-2)

wherein:

15 R represents a hydrogen or halogen atom; a straight- or branched-chain alkyl or alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a cycloalkyl group containing from 3 to 6 carbon atoms optionally substituted by one or more groups  $R^5$ , one or more halogen atoms or a group  $-CO_2R^3$ ; or a group selected from  $-CO_2R^3$ ,  $-COR^5$ , cyano, nitro,  $-CONR^3R^4$  and  $-S(O)_kR^{13}$ ,

20  $R_1$  represents a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a cycloalkyl group containing from three to six carbon atoms optionally substituted by one or more groups  $R^5$  or one or more halogen atoms;

25  $R^2$  represents a halogen atom; a straight- or branched-chain alkyl, alkenyl or alkynyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; a straight- or branched-chain alkyl group containing up to six carbon atoms which is substituted by one or more groups  $-OR^5$ ; or a group selected from nitro, cyano,  $-CO_2R^5$ ,  $-S(O)_pR^6$ ,  $-O(CH_2)_mOR^5$ ,  $-COR^5$ ,  $-NR^{11}R^{12}$ ,  $-N(R^8)SO_2R^7$ ,  $-N(R^8)CO_2R^7$ ,  $-OR^5$ ,  $-OSO_2R^7$ ,  $-SO_2NR^3R^4$ ,  $-CONR^3R^4$ ,  $-CSNR^3R^4$ ,  $-(CR^9R^{10})_l-S(O)_qR^7$  and  $-SF_5$ ; or two groups  $R^2$ , on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, form a 5

to 7 membered saturated or unsaturated heterocyclic ring containing up to three ring heteroatoms selected from nitrogen, oxygen and sulfur, which ring is optionally substituted by one or more groups selected from halogen, nitro,  $-S(O)_pR^{13}$ ,  $C_{1-4}$  alkyl,  $C_{1-4}$  alkoxy,  $C_{1-4}$  haloalkyl,  $C_{1-4}$  haloalkoxy,  $=O$  (or a 5- or 6-membered cyclic acetal thereof) and  $=NO-R_3$ , it being understood that a sulphur atom, where present in the ring, may be in the form of a group  $-SO-$  or  $-SO_2-$ ;

n represents an integer from one to five: when n is greater than one the groups  $R^2$  may be the same or different;

$R^3$  and  $R^4$  each independently represent a hydrogen atom, or a straight- or branched chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

$R^5$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; or a straight- or branched-chain alkenyl or alkynyl group containing from two to six (preferably from three to six) carbon atoms which is optionally substituted by one or more halogen atoms;

$R^6$  and  $R^7$ , which may be the same or different, each represent  $R^5$ , or phenyl optionally substituted by from one to five groups which may be the same or different selected from a halogen atom, a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms; nitro, cyano,  $-CO_2R^5$ ,  $-S(O)_pR^{13}$ ,  $-NR^{11}R^{12}$ ,  $-OR^5$  and  $-CONR^3R^4$ ;

$R^8$ ,  $R^9$ , and  $R^{10}$  each represent a hydrogen atom or  $R^6$ ;

$R^{11}$  and  $R^{12}$  each present hydrogen or  $R^5$ ;

$R^{13}$  represents a straight- or branched-chain alkyl group containing up to six carbon atoms which is optionally substituted by one or more halogen atoms;

k, p and q independently represent the values zero, one or two;

m represents one, two or three;

t represents an integer from one to four; when t is greater than one, the groups  $R^9$  and  $R^{10}$  may be the same or different;

or an agriculturally acceptable salt or metal complex thereof.

In certain cases, the groups R to  $R^{13}$  may give rise to optical and/or

stereoisomerism. All such forms are embraced by the present invention.

By the term "agriculturally acceptable salts" is meant salts the cations or anions of which are known and accepted in the art for the formation of salts for agricultural or horticultural use. Preferably the salts are water soluble. Suitable acid addition salts, formed by compounds of formula (I) containing an amino group, include salts with inorganic acids, for example, hydrochlorides, sulphates, phosphates and nitrates, and salts with organic acids, for example, acetic acid. Suitable salts formed by compounds of formula (I) which are acidic, i.e. compounds containing one or more carboxy groups, with bases include alkali metal (e.g. sodium and potassium) salts, alkaline earth metal (e.g. calcium and magnesium) salts, ammonium and amine (e.g. diethanolamine, triethanolamine, octylamine, dioctylmethylamine and morpholine) salts.

In the description unless otherwise specified, 'halogen' means a fluorine, chlorine, bromine or iodine atom.

Compounds of formula (I) wherein A represents (A-1) are preferred.

The phenyl ring of the compounds of formula (I) is preferably 2,4-disubstituted 2,3-disubstituted or 2,3,4-trisubstituted.

Compounds of formula (I) in which R represents hydrogen or  $-\text{CO}_2\text{R}^3$  wherein  $\text{R}^3$  represents a straight- or branched chain alkyl group containing up to three carbon atoms; and  $\text{R}^1$  represents cyclopropyl are preferred.

Compounds of formula (I) in which  $\text{R}^2$  represents a halogen atom; a straight- or branched-chain alkyl group containing up to three carbon atoms which is optionally substituted by one or more halogen atoms;  $-\text{S}(\text{O})_p\text{R}^6$ ;  $-\text{OR}^5$  or  $-\text{CH}_2\text{S}(\text{O})_q\text{R}^7$ ; wherein  $\text{R}^5$ ,  $\text{R}^6$  and  $\text{R}^7$  each represent the same or different optionally halogenated methyl or ethyl groups are preferred.

A preferred class of compounds of formula (I) wherein A represents (A-1) are those wherein:

R is hydrogen or  $-\text{CO}_2\text{E}$ ;

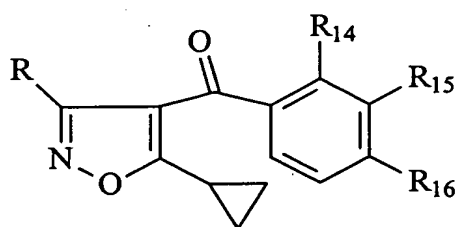
$\text{R}^1$  is cyclopropyl;

and two groups  $\text{R}^2$ , on adjacent carbon atoms of the phenyl ring may, together with the carbon atoms to which they are attached, combine to form a 5 or 6

membered saturated or unsaturated heterocyclic ring which is fused to the 2,3 or 3,4 positions of the benzoyl ring; wherein the heterocyclic ring contains two hetero atoms selected from sulphur and oxygen which are located at the 2 and 3, or 3 and 4 positions of the benzoyl ring; and in which the 4-substituent of the benzoyl ring is halogen or  $S(O)_pMe$ , or the 2-substituent of the benzoyl ring is methyl,  $S(O)_pMe$  or  $-CH_2S(O)_qMe$  respectively; and optionally the heterocyclic ring may be substituted by one or more halogen atoms.

A more preferred class of compounds of formula (I) are those wherein A represents (A-1); R is hydrogen or  $-CO_2Et$ ;  $R^1$  is cyclopropyl;  $R^2$  is a halogen atom or a group selected from  $-CF_3$ , Me, Et,  $-S(O)_pMe$ ,  $-CH_2S(O)_qMe$  and optionally halogenated methoxy or ethoxy; and n is two or three.

A yet more preferred class of compounds of formula (I) are those having the formula (Ia):



(Ia)

wherein:

R is hydrogen or  $-CO_2Et$ ;

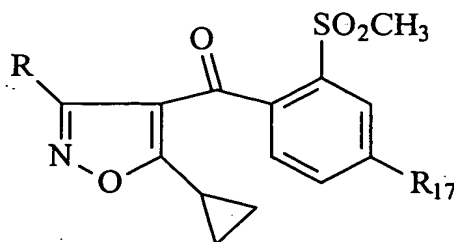
$R^{14}$  is selected from  $-S(O)_pMe$ , Me, Et, a chlorine, bromine or fluorine atom, methoxy, ethoxy and  $-CH_2S(O)_qMe$ ;

$R^{15}$  is selected from a hydrogen atom, a chlorine, bromine or fluorine atom, methoxy, ethoxy and  $-S(O)_pMe$ ;

$R^{16}$  is selected from a hydrogen atom, a chlorine, bromine or fluorine atom, methoxy and  $CF_3$ ;

and wherein at least one of  $R^{15}$  and  $R^{16}$  is other than hydrogen.

An especially preferred class of compounds of formula (I) have the formula (Ib):



(Ib)

wherein  $R^{17}$  is chlorine, bromine or trifluoromethyl; and

$R$  is hydrogen or  $CO_2Et$ .

The following compounds of formula (I) are among the most preferred for use in the present invention:

- 5-cyclopropyl-4-[2-chloro-3-ethoxy-4-(ethylsulphonyl)benzoyl]isoxazole;
- 4-(4-chloro-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;
- 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole;
- 4-(4-bromo-2-methylsulphonylbenzoyl)-5-cyclopropylisoxazole;
- 5-cyclopropyl-4-[4-fluoro-3-methoxy-2-(methylsulphonyl)benzoyl]isoxazole;
- 4-(4-bromo-2-methylsulphonylmethylbenzoyl)-5-cyclopropylisoxazole;
- ethyl 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole-3-carboxylate;
- 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)-3-methylthioisoxazole.

The most preferred compound is 5-cyclopropyl-4-(2-methylsulphonyl-4-trifluoromethylbenzoyl)isoxazole (isoxaflutole).



Procedures for preparing isoxazoles of formula (I) are as described in European Patent Publication Nos. 0418175, 0487357, 0527036 and 0560482.

The method of the invention can be used on genetically modified crops.

By genetically modified crop is understood those crops which have been made tolerant towards herbicides by conventual sowing and cultivation methods or genetic engineering methods.

According to a further feature of the present invention, there are provided compositions suitable for herbicidal use in the method of the invention comprising one or more of the 4-benzoylisoxazoles of formula (I) or an agriculturally acceptable salt or metal complex thereof (which may be encapsulated as hereinbefore described), in association with, and preferably homogeneously dispersed in, one or more compatible agriculturally-acceptable diluents or carrier and/or surface active agents [i.e. diluents or carriers and/or surface active agents of the type generally accepted in the art as being suitable for use on herbicidal compositions and which are compatible with compounds of formula (I)]. The term "homogeneously dispersed" is used to include compositions in which the compounds of formula (I) are dissolved in other components. The term "herbicidal compositions" is used in a broad sense to include not only compositions which are ready for use as herbicides but also concentrates which must be diluted before use. Preferably, the compositions contain from 0.05 to 90% by weight of one or more compounds of formula (I).

The herbicidal compositions may contain both a diluent or carrier and surface-active (e.g. wetting, dispersing, or emulsifying) agent. Surface-active agents which may be present in herbicidal compositions of the present invention may be of the ionic or non-ionic types, for example sulphoricinoleates, quaternary ammonium derivatives, products based on condensates of ethylene oxide with alkyl and polyaryl phenols, e.g. nonyl- or octyl-phenols, tristeryl phenols, condensates of ethylene oxide with alcohols, or carboxylic acid esters of anyhydrosorbitols which have been rendered soluble by etherification of the free hydroxy groups by condensation with ethylene oxide, alkali and alkaline earth metal salts of sulphuric acid esters and sulphonic acids such as dinonyl- and dioctyl- sodium sulphonosuccinates and alkali and alkaline earth metal salts of high molecular weight

sulphonic acid derivatives such as sodium and calcium lignosulphonates and sodium and calcium alkylbenzene sulphonates.

Suitably, the herbicidal compositions according to the present invention may comprise up to 10% by weight, e.g. from 0.05% to 10% by weight, of surface-active agent but, if desired, herbicidal compositions according to the present invention may  
5 comprise higher portions of surface-active agent, for example up to 15% by weight in liquid emulsifiable suspension concentrates and up to 25% by weight in liquid water soluble concentrates.

Examples of suitable solid diluents or carriers are aluminium silicate,  
10 microfine silicon dioxide, talc, chalk, calcined magnesia, kieselguhr, tricalcium phosphate, powdered cork, adsorbent carbon black and clays such as kaolin, attapulgite, diatomaceous earth, mica, alumina oxide, titanium oxide and bentonite. The solid compositions (which may take the form of dusts, granules or wettable  
15 powders) are preferably prepared by grinding the compounds of formula (I) with solid diluents or by impregnating the solid diluents or carriers with solutions of the compounds of formula (I) in volatile solvents, evaporating the solvents and if  
20 necessary, grinding the products so as to obtain powders. Granular formulations may be prepared by absorbing the compounds of formula (I) dissolved in suitable solvents, (which may, if desired, be volatile) onto the solid diluents or carriers in  
granular form and, if desired, evaporating the solvents, or by granulating  
compositions in powder form obtained as described above. Solid herbicidal  
compositions, particularly wettable powders and granules, may contain wetting or  
dispersing agents (for example of the types described above), which may also, when  
solid, serve as diluents or carriers.

25 Liquid compositions according to the invention may take the form of aqueous, organic or aqueous-organic solutions, suspensions and emulsions which may incorporate a surface-active agent. Suitable liquid diluents for incorporation in the liquid compositions include water, glycols, glycol ethers, tetrahydrofurfuryl  
alcohol, acetophenone, cyclohexanone, isophorone, alkyl pyrrolidones,  
30 butyrolactone, chlorinated toluene, xylene, mineral, animal and vegetable oils, esterified vegetable oils and light aromatic and naphthenic fractions of petroleum

(and mixtures of these diluents). Surface-active agents, which may be present in the liquid compositions, may be ionic or non-ionic (for example of the types described above) and may, when liquid, also serve as diluents or carriers.

5 Powders, dispersible granules and liquid compositions in the form of concentrates may be diluted with water or other suitable diluents, for example mineral or vegetable oils, particularly in the case of liquid concentrates in which the diluent or carrier is an oil, to give compositions ready for use.

10 When desired, liquid compositions of the compounds of formula (I) may be used in the form of self-emulsifying concentrates containing the active substances dissolved in the emulsifying agents or in solvents containing emulsifying agents compatible with the active substances, the simple addition of such concentrates to water producing compositions ready for use.

Liquid concentrates in which the diluent or carrier is an oil may be used without further dilution using the electrostatic spray technique.

15 Herbicidal compositions according to the present invention may also contain, if desired, conventional adjuvants such as adhesives, protective colloids, thickeners, penetrating agents, spreading agents, stabilisers, buffers, sequestering agents, anti-caking agents, colouring agents and corrosion inhibitors. These adjuvants may also serve as carriers or diluents.

20 Unless otherwise specified, the following percentages are by weight. Preferred herbicidal compositions according to the present invention are encapsulations containing water dispersible granules which comprise from 1 to 90%, e.g. 25 to 75% of one or more compounds of formula (I), from 1 to 15%, e.g. 2 to 10%, of surface-active agent and from 5 to 95%, e.g. 20 to 60%, of solid diluent, e.g. 25 clay, granulated with the addition of water to form a paste and then dried;

aqueous suspension concentrates which comprise from 5 to 70% of one or more compounds of formula (I), from 2 to 10% of surface-active agent, from 0.1 to 5% of thickener and from 15 to 87.9% of water;

30 wettable powders which comprise from 5 to 90% of one or more compounds of formula (I), from 2 to 10% of surface-active agent and from 8 to 88% of solid diluent or carrier;

water soluble or water dispersible powders which comprise from 5 to 90% of one or more compounds of formula (I), from 2 to 40% of sodium carbonate and from 0 to 88% of solid diluent;

liquid water soluble concentrates which comprise from 5 to 50%; e.g. 10 to 30% of one or more compounds of formula (I), from 0 to 25% of surface-active agent and from 10 to 90%, e.g. 45 to 85%, of water miscible solvent, e.g. triethylene glycol, or a mixture of water-miscible solvent and water;

liquid emulsifiable suspension concentrates which comprise from 5 to 70% of one or more compounds of formula (I), from 5 to 15% of surface-active agent, from 0.1 to 5% of thickener and from 10 to 84% of organic solvent, e.g. mineral oil; and

emulsifiable concentrates which comprise 0.05 to 90%, and preferably from 1 to 60% of one or more compounds of formula (I), from 0.01 to 10%, and preferably from 1 to 10%, of surface-active agent and from 9.99 to 99.94%, and preferably from 39 to 98.99%, of organic solvent.

The water dispersible granules comprising isoxazoles of formula (I) whose apparent density was 0.25-0.75, have a particle size of generally 10-2000 $\mu$ m, preferably 300-1500 $\mu$ m.

Herbicidal compositions according to the present invention may also comprise the compounds of formula (I) in association with, and preferably homogeneously dispersed in, one or more other pesticidally active compounds and, if desired, one or more compatible pesticidally acceptable diluents or carriers, surface-active agents and conventional adjuvants as hereinbefore described.

Examples of other pesticidally active compounds which may be included in, or used in conjunction with, the herbicidal compositions of the present invention include herbicides, for example to increase the range of weed species controlled for example acetochlor,

alachlor [2-chloro-2,6'-diethyl-N-(methoxy-methyl)-acetanilide], atrazine [2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine], bromoxynil [3,5-dibromo-4-hydroxybenzonitrile], chlortoluron [N'-(3-chloro-4-methylphenyl)-N,N-dimethylurea], cyanazine [2-chloro-4-(1-cyano-1-methylethylamino)-6-ethylamino-1,3,5-triazine], 2,4-D [2,4-dichlorophenoxy-acetic acid], dicamba [3,6-dichloro-2-

methoxybenzoic acid], difenzoquat [1,2-diethyl-3,5-diphenyl-pyrazolium salts], dimethanamid, flampropmethyl [methyl N-2-(N-benzoyl-3-chloro-4-fluoroanilino)-propionate], flufenacet, fluometron [N'-(3-trifluoro-methylphenyl)-N,N-dimethylurea], glyphosate, glufosinate, isoproturon [N'-(4-isopropylphenyl)-N,N-dimethylurea], metolachlor, metribuzin, insecticides, e.g. synthetic pyrethroid, e.g. permethrin and cypermethrin, fipronil and fungicides, e.g. carbamates, e.g. methyl N-(1-butyl-carbamoyl-benzimidazol-2-yl)carbamate, and triazoles e.g. 1-(4-chlorophenoxy)-3,3-dimethyl-1-(1,2,4-triazol-1-yl)-butan-2-one.

Pesticidally active compounds and other biologically active materials which may be included in, or used in conjunction with, the herbicidal compositions of the present invention, for example those hereinbefore mentioned, and which are acids, may, if desired, be utilized in the form of conventional derivatives, for example alkali metal and amine salts and esters.

The following Examples illustrate herbicidal compositions which may be used in the present invention. The Active Ingredient listed in the following examples refers to a compound of general formula (I).

**Example C1:**

An emulsifiable concentrate is formed from:

Active ingredient	20% w/v
N-Methylpyrrolidinone (NMP)	25% w/v
Calcium dodecylbenzenesulphonate (CaDDBS)	70% 4% w/v
Nonylphenol ethylene oxide propylene oxide condensate (NPEOPO)	4% w/v
Aromatic solvent	to 100 volumes
by stirring NMP, active ingredient (Compound 1), CaDDBS,	

NPEOPO and Aromatic solvent until a clear solution is formed, and adjusting to volume with Aromatic solvent.

**Example C2**

A wettable powder is formed from:

Active Ingredient	50% w/w
Sodium dodecylbenzenesulphonate	3% w/w
Sodium methyl oleoyl taurate	5% w/w
Sodium polycarboxylate	1% w/w
Sodium polycarboxylate	1% w/w
Microfine silicon dioxide	3% w/w
China clay	38% w/w

by blending the above ingredients together and grinding the mixture in an air jet mill.

**Example C3**

A suspension concentrate is formed from:

Active Ingredient	50% w/v
Antifreeze (Propylene glycol)	50% w/v
Ethoxylated tristyrylphenol phosphate	0.5% w/v
Nonyl phenol 9 mole ethoxylate	0.05% w/v
Sodium polycarboxylate	0.02% w/v
Attaclay	1.5% w/v
Antifoam	0.003% w/v
Water	to 100 volumes

by stirring the above ingredients together and milling in a bead mill.

**Example C4**

A water dispersible granule is formed from:

Active ingredient	50% w/w
Sodium dodecylbenzenesulphonate	3% w/w
Sodium methyl oleoyl taurate	5% w/w

Sodium polycarboxylate	1% w/w
Binder (Sodium lignosulphonate)	8% w/w
china clay	30% w/w
Microfine silicon dioxide	3% w/w

5 by blending the above ingredients together, grinding the mixture in an air jet mill and granulating by addition of water in a suitable granulation plant (e.g. Fluid bed drier) and drying. Optionally the active ingredient may be ground either on its own or admixed with some or all of the other ingredients.

10 The following non-limiting Example illustrates the invention.

### EXAMPLE 1

15 Glasshouse experiment showing unexpected enhancement of biological activity on weeds following application of compound (isoxaflutole).

7 x 7 cm<sup>2</sup> pots were filled with a non-sterile loam soil. Weed seeds (*Amaranthus retroflexus*, *Echinochloa crus galli* and *Setaria viridis*) were placed in three separate shallow wells with a maize seed (Pioneer 3394) inserted to a depth of 4 cm in between in each pot and the seeds lightly covered with soil.

20 A 1 ml solution of technical isoxaflutole, suitably diluted to give a dose equivalent to 6.25, 12.5, 25, 50 and 100 /ha in acetonitrile, was pipetted evenly on the soil surface of pots (replicated 10 times), according to the following regime.

On day 1, a set of 5 pots were treated with 5 dose rates (6.25-100 g/ha) and set aside in the glasshouse. At the same time, on day 1 a second set of 4 pots were  
25 treated with 4 dose rates (treatment A : 6.25-50 g/ha) along with a third set of 3 pots treated with 3 dose rates (treatment B : 6.25-25 g/ha). On day 2 each of the second and third pots were treated again the pots receiving identical treatments A and B and the second set of pots set aside in the glasshouse. On day 3 each of the third set of pots were treated again, the pots receiving identical treatment B. On day 4 each of  
30 the third set of pots were treated, the pots receiving identical treatment B and the pots set aside in the glasshouse.

The pots were maintained in a glasshouse, with overhead watering (3 x daily) and supplementary lighting. Visual assessment of % reduction, compared to untreated control plants was recorded 14 days after treatment. The results (average of 10 replicates) are shown in Table 1, where the dosing regime refers to the number of daily applications followed by the dose of compound on each day.

**TABLE 1**

**% Damage 14 DAT: Comparison of Single vs. Multiple Application**

% Damage				
Dosing Regime	Amare	Echcg	Setvi	Maize (P3394)
2 x 6.25	34	18	18	0
1 x 12.5	0	0	0	0
4 x 6.25	80	97	75	0
2 x 12.5	46	68	42	0
1 x 25	8	32	10	0
4 x 12.5	79	100	91	6
2 x 25	42	70	44	0
1 x 50	50	84	24	2
4 x 25	90	100	77	6
2 x 50	78	96	66	10
1 x 100	79	94	59	8

As can be seen in Table 1, the efficacy of the herbicide on key weed species is improved by 2 applications separated by 1 day and surprisingly even more enhanced by 4 applications separated by 3 days, compared to a single application of the same total dose of compound. Repeat applications did not appear to have any impact on maize phytotoxicity.

## EXAMPLE 2

A 30% aqueous solution of Gerol was prepared and 67g added to 80g of a



suspension of isoxaflutole (250g/L) in water containing a dispersant (sodium polynaphthalene sulphate to maintain the suspension) with stirring. Powdered calcium carbonate (1g) was added and the resultant mixture sonicated with ultrasound before addition of polysiloxane oil (600 ml) to give an emulsion. Acetic acid (3ml) was added and stirring continued for 2 hours. The stirring was stopped and the upper layer decanted from the precipitate. This solid was filtered off, washed with water and dried to give microparticles of encapsulated isoxaflutole. The encapsulated material may be formulated using methods hereinbefore described.

10 Gerol: Diethyleneglycol-ethyleneglycol-isophthalic acid-sodium 5-sulfoisophthalate-terephthalic acid-triethylene glycol copolymer.

## EXAMPLE 3

Maize is sown and then grown up in area where weeds and weed seeds are present. The weeds are a selection from: Alopecurus myosuroides, Avena fatua, Digitaria sanguinalis, Echinochloa crus-galli, Eleusine indica, Lolium multiflorum, Setaria viridis, Sorghum halepense, Cyperus esculentus, Cyperus iria, Cyperus rotundus, Eleocharis acicularis, Abutilon theophrasti, Amaranthus retroflexus, Bidens pilosa, Chenopodium album, Galium aparine, Ipomoea purpurea, Lamium purpureum, Matricaria inodora, Sesbania exalta, Sinapis arvensis, Solanum nigrum, Stellaria media, Veronica hederifolia, Veronica persica, Viola arvensis, and Xanthium strumarium.

After one week from sowing the maize, isoxazole is sprayed as an encapsulated formulation as hereinbefore described at a rate of 105 g/ha of herbicidally active compound, the weight ratio of isoxazole : copolyester being 1 : 10. The amounts of isoxazole of formula (I) and DKN were measured after 4 days in a 5cm deep soil core around the seed or seedling. The weight ratio of isoxazole : DKN was found to be 1.

The activity of the isoxazole on the crop and weeds was observed after 3 weeks, and found to be equal to 2% and 95% respectively.

A similar application in similar soil conditions without the copolyester provided a weight ratio of isoxazole : DKN of 0.1 and herbicidal activity on both crop and weeds was 15% and 95% respectively.

## EXAMPLE 4

Maize is sown in an area where weeds and weed seeds of the species Setaria viridis are present. Isoxazole is sprayed on the soil surface as an encapsulated formulation at a rate of 105g herbicidally active compound per hectare. The activity of the isoxazole on maize and Setaria viridis was observed 6, 11, 14 and 17 days after treatment (DAT).

Activity of 4-benzoylisoxazole on Setaria viridis

Application rate g/ha	Formulation	DAT			
		6	11	14	17
105	encapsulated	0	40	70	90
	WG	10	50	75	100

#### Activity of 4-benzoylisoxazole on Maize

Application rate g/ha	Formulation	DAT			
		6	11	14	17
105	encapsulated	0	0	3	8
	WG	0	5	7	10

Activity on Setaria viridis by the encapsulated formulation was equal to that of the WG formulation. The encapsulated formulation decreased phytotoxicity on maize by 20-30% compared to the WG formulation.

#### EXAMPLE 5

Maize is sown and grown up in an area where weeds and weed seedlings are present. The weeds are Amaranthus retroflexus, Echinochloa crus-galli and Setaria viridis. Solutions of isoxazole in acetonitrile were applied directly to the soil surface at dose rates of 100, 50 and 25g/ha to plots A, B and C respectively. 1 DAT dose rates of 50 and 25g/ha were applied to plots B and C respectively, 2 and 3 DAT a dose rate of 25g/ha was applied to plot C.

Activity of the isoxazole on maize and the weed species was observed 14 DAT.

Dosing regime	AMARE	ECHCG	SETVI	Maize
4 x 25g/ha	90	100	77	6
2 x 50g/ha	78	96	66	10
1 x 100g/ha	79	94	59	8

5 Maintaining the isoxazole in the surface of the soil within the area containing weed seeds and seedlings, by sequential applications of low dose rates of isoxazole, provides improved activity on weeds.

## CLAIMS

1. A method for controlling the growth of weeds at a locus in a solid growing medium which comprises treating the locus with a composition comprising an isoxazole herbicide to provide progressive or sequential delivery or release of isoxazole herbicide into the surface layer of the medium.

2. A method according to claim 1 in which the growing medium is soil.

3. A method according to claim 1 or 2 in which the locus is a crop-growing locus.

4. A method according to claim 1, 2 or 3 in which the surface layer of the medium is from the surface to a depth of 10 cm.

5. A method according to any one of the preceding claims which comprises applying to the locus sequential low doses of isoxazole herbicide.

6. A method according to any one of claims 1 to 4 which comprises treating the locus with a delayed release composition comprising the isoxazole herbicide.

7. A method according to claim 6 in which the delayed release composition comprises an encapsulated composition.

8. A method according to claim 6 or 7 in which an encapsulated isoxazole is used, comprising an isoxazole derivative encapsulated with a solid film comprising an inert material itself having no substantial herbicidal activity.

9. A method according to claim 8 in which granules of an isoxazole derivate from 0.1 to 50  $\mu\text{m}$  in size are used.

10. A method according to any one of the preceding claims in which the isoxazole derivative is of general formula I as hereinbefore defined.

11. A delayed release composition comprising an isoxazole herbicide.

5

12. A method according to claim 1 substantially as hereinbefore described in Example 1.

13. A delayed release composition according to claim 9 substantially as  
10 hereinbefore described.

ABSTRACT

5       The invention provides a method for controlling the growth of weeds at a locus in a solid growing medium which comprises treating the locus with a composition comprising an isoxazole herbicide to provide progressive or sequential delivery or release of isoxazole herbicide into the surface layer of the medium.

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